Microgravity Studies of Liquid-Liquid Phase Transitions in Undercooled Alumina-Yttria Melts

Richard Weber and Paul Nordine
Containerless Research, Inc.
Evanston, IL 60201-3149

Phone: (847) 467-2678
email: weber@containerless.com

Introduction and Background

The scientific objective of this research is to increase the fundamental knowledge base for liquid-phase processing of technologically important oxide materials. The experimental objective is to define precise conditions and hardware requirements for microgravity flight experiments to test and expand the experimental hypotheses that:

1. Liquid-liquid phase transition can occur in undercooled melts by a diffusionless transformation.

2. Onset of the liquid-liquid phase transition is accompanied by a large change in the temperature dependence of viscosity.

Recent experiments on undercooled YAG ($Y_3Al_5O_{12}$)-composition liquid demonstrated a large departure from an Arrhenian temperature dependance of viscosity.[1] Fibers were pulled from melts undercooled by *ca.* 600° indicating that the viscosity is on the order of 1000 Poise (100 Pa•s) at 1600 K.[1] This value of viscosity is 500 times greater than that obtained by extrapolation of data for temperatures close to the melting point of YAG.[2] YAG is known to be a very "fragile" glass former[3] and the recent results suggest that the onset of the highly non-Arrhenian viscosity-temperature relationship occurs at a temperature about 600° below the equilibrium melting point.

Additional experimental results on alumina-yttria melts richer in aluminum than the YAG-composition suggest that a liquid phase transition occurs in the undercooled liquids at rates inconsistent with diffusion limited separation processes.[4] This raises the possibility that a congruent liquid phase transition occurs in the YAG composition liquid. The phase transition results in the large change in melt viscosity which is the subject of this investigation.

Experimental Approach

The research focuses on the study of undercooled pseudo-binary alumina-yttria melts. Although these highly stable oxides exhibit only very small oxygen composition changes some properties of oxide melts are very sensitive to the ambient oxygen pressure[5] which must be controlled in order to investigate liquid oxides. A further property of this system is that the melts are partially transparent so that the interior of the specimen can be observed *in-situ* to study the onset and kinetics of phase transition, nucleation and growth.

Long term microgravity conditions are needed to meet all of the requirements for detailed investigation of the phase transition phenomena:

1. Access to undercooled melts with a high degree of control of melt purity, quiescence, and the ambient oxygen partial pressure.

2. Gas-liquid equilibration at the extremely slow, diffusion limited rates allowing property measurements on a time scale that is small compared to the gas liquid equilibration time.

3. Elimination of gravity effects on buoyancy-driven convection, phase growth rate, settling time, and coalescence.

4. Fully-controlled synthesis of materials for subsequent examination and analysis.

Earth-based research is being performed under conditions that meet some of these constraints to provide the design basis for microgravity experiments. Containerless experiments using aero-acoustic (AAL)[6] and electrostatic (ESL)[7] levitation techniques are being used to achieve precise control of melt chemistry, access highly undercooled melts and glassy states, and control the effects of interaction with container walls on the evolution of morphology. Microgravity experiments will obtain the control of thermal gradients, density-driven segregation, and especially of fluid flow required to test the experimental hypotheses.

Results/Plans

Preliminary experiments to demonstrate levitation and melting of YAG were recently performed using the ESL at NASA Marshall Space Flight Center (MSFC). Detailed experiments on molten oxide compositions, using CO_2 laser beam heating, are scheduled at MSFC in June 1998.

References

1. J.K.R. Weber, J.J. Felten, B. Cho, and P.C. Nordine. "Glass Fibers of Pure and Erbium or Neodymium-doped Yttria-alumina Compositions," *Nature*, in press.

2. V.J. Fratello and C.D. Brandle. "Physical Properties of a $Y_3Al_5O_{12}$ Melt," *J. Cryst. Growth*, 128, 1006-1010 (1993).

3. D. Massiot, F. Taulelle, and J.P. Coutures. "Structural Diagnostic of High Temperature Liquid Phases by ^{27}Al NMR," *Colloque de Phys.*, C5, 425-431 (1990).

4. S. Aasland and P.F. McMillan. "Density-driven Liquid-liquid Phase Separation in the Al_2O_3-Y_2O_3 System," *Nature*, 369, 633 (1994).

5. J.K.R. Weber, S. Krishnan, and P.C. Nordine. "Effects of Melt Chemistry on the Spectral Absorption Coefficient of Liquid Aluminum Oxide," *J. Am. Ceram. Soc.*, 78, 3067-3071 (1995).

6. J.K.R. Weber, D.S. Hampton, D.R. Merkley, C.A. Rey, M.M. Zatarski, and P.C. Nordine. "Aero-acoustic Levitation - A Method for Containerless Liquid-phase Processing at High Temperatures," *Rev. Sci. Instrum.*, 65, 456-465 (1994).

7. A.J. Rulison, J.L. Watkins, and B. Zambrano. "Electrostatic Containerless Processing System," *Rev. Sci. Instrum.*, 68, 2856-2863 (1997).